

REMARKS

The applicant respectfully requests reconsideration in view of the following remarks.

The applicant has rewritten claim 11 into independent form by incorporating claim 1 into claim 11. The applicant has changed the dependencies of claims 2-6 and 8-10. No new matter has been added.

Claims 1-6, 8-17, 19, 20 and 22 are rejected under 35 U.S.C. 103(a) as being unpatentable over Frauenkron (US Patent 6,562,971) in view of Ashina (European Patent Publication 0,130,407) and Ogawa (US Patent 6,350,874). The applicant respectfully traverses this rejection.

The present invention relates to a preparing triethylenediamine (TEDA) **which is a cyclic tert.-amine** by reaction of ethylenediamine (EDA) and/or piperazine (PIP) in the presence of a crystalline aluminosilicate catalyst, wherein a shaped body whose cutting hardness has been increased beforehand using a method of increasing the cutting hardness of a shaped body comprising a crystalline aluminosilicate having an $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 10:1 to 1200:1, wherein the shaped body comprises a binder selected from among oxides of silicon and/or zirconium and is treated with a gas consisting of water vapor at from 100 to 600°C and an absolute pressure of from 0.1 to 10 bar for a period of at least 20 hours and the shaped body has been calcined at from 100 to 600°C before the treatment with water vapor is used as the catalyst (see the specification at page 1, lines 1-8, and claim 11).

In the last response, the applicant pointed out that Frauenkron taught away from the claimed molar ratio.

Frauenkron discloses at col. 8, lines 10-39:

The zeolite employed as catalyst in the process according to the invention for the preparation of TEDA has a skeletal structure which principally consists of silicon dioxide (SiO_2).

Besides silicon dioxide, the zeolite may also contain traces of other divalent, trivalent or tetravalent metals **M (oxidation state of the metals II, III or IV)** in the form of metal oxides $\text{M}_{2/z} \text{O}$ ($z=2, 3$ or 4), where the molar ratio of silicon dioxide and the metal oxide, $\text{SiO}_2 / \text{M}_{2/z} \text{O}$ (this molar ratio is also known as the 'modulus') is greater than 10 for tetravalent metals ($z=4$), greater than 100 for divalent and trivalent metals ($z=2$ or 3) and greater than 1400 for $\text{M}=\text{Al}(z=3)$.

The zeolite catalysts preferably used in the process according to the invention have, for $\text{M}=\text{Al}$, an $\text{SiO}_2 / \text{M}_2 \text{O}_3$ molar ratio of from greater than 1400 to 40,000:1, in particular from greater than 1400 to 5000:1, for $\text{M}=\text{metal}$ in oxidation state II or $\text{M}=\text{two or more metals}$ in oxidation state II, have an SiO_2 / MO molar ratio of from greater than 100 to 40,000:1, in particular from greater than 200 to 5000:1, for $\text{M}=\text{metal}$ in oxidation state III or $\text{M}=\text{two or more metals}$ in oxidation state III, have an $\text{SiO}_2 / \text{M}_2 \text{O}_3$ molar ratio of from greater than 100 to 40,000:1, in particular from greater than 200 to 5000:1, and for $\text{M}=\text{metal}$ in oxidation state IV or $\text{M}=\text{two or more metals}$ in oxidation state IV, have an $\text{SiO}_2 / \text{MO}_2$ molar ratio of from greater than 10 to 40,000:1, in particular from greater than 25 to 5000:1. (emphasis added)

For Al_2O_3 , Frauenkron teaches that the molar ratio of $\text{SiO}_2 / \text{Al}_2\text{O}_3$ is from greater than 1400 to 40,000:1, in particular from greater than 1400 to 5000:1. Frauenkron's minimum ratio is **greater than 1400** while the applicant's claimed maximum ratio is 1200. Therefore, Frauenkron teaches away from the applicant's claimed invention ($\text{SiO}_2 / \text{Al}_2\text{O}_3$ molar ratio of 10:1 to 1200:1) and teaches to have at least a molar ratio of $\text{SiO}_2 / \text{Al}_2\text{O}_3$ being greater than 1400.

As the Examiner has recognized at page 3 of the office action, Frauenkron does neither disclose a shaped catalyst body which is treated with a gas consisting of water vapor beforehand the reaction, nor a shaped catalyst body having an $\text{SiO}_2 / \text{Al}_2\text{O}_3$ molar ratio of 10:1 to 1200:1.

The Examiner has relied upon Ogawa for the teaching of the claimed ratio. However,

Ogawa also does not teach a shaped catalyst body is treated according to claim 11 before it is used as catalyst.

Moreover, Frauenkron and Ogawa are silent in view of a process, wherein a shaped body whose cutting hardness has been increased beforehand the reaction according to claim 11 is used as a catalyst.

The Examiner states that Ashina discloses the use of a zeolite as a catalyst for vapor phase reactions which includes first steaming the zeolite with steam to increase the results of the reaction (see page 3 of the office action). This statement does not consider the whole disclosure of Ashina.

Ashina describes a process for producing dimethylamine (DMA) by a gas phase reaction of ammonia and methanol over a catalyst of a zeolite, wherein said catalyst is previously and separately from said reaction of ammonia and methanol contacted with an atmosphere comprising steam at a temperature of from 250 to 700°C (see claim 11 and the abstract). The technical effect of a process described by Ashina is the improvement of the DMA selectivity in preference to mono- and trimethylamines (see the abstract and examples of Ashina).

Therefore, Ashina is drawn to a totally different technical field. Ashina is drawn to the production of DMA by a condensation reaction of methanol (CH_3OH) and ammonia (NH_3). The technical effect achieved by the teaching of Ashina is selectivity improvement of a non-cyclic sec.-amine in preference to primary amines and tertiary amines.

The applicant's reaction is not a condensation reaction. Moreover, it is surprising that in contrary to Ashina a selectivity improvement of a cyclic tertiary-amine is achieved. By the process of Ashina a selectivity improvement of a non-cyclic secondary-amine is achieved and

NOT a cyclic amine. A process producing dimethylamine as described in Ashina is completely different to a process for producing triethylenediamine (TEDA) according to the present application.

The applicant has enclosed evidence that these processes are not interchangeable. The first additional prior art reference (citation from Ullmann' s Encyclopedia of Industrial Chemistry; page 5 in connection with the production of lower alkylamines such as DMA), which discloses the production of lower alkylamines is generally carried out at a pressure of 5 to 250 bar, a temperature of 100 to 250°C and a two- to eightfold excess of ammonia versus the alcohol (see right-hand column, 1st and last paragraph). Furthermore, the reaction can be carried out either in the liquid-phase or in the gas-phase.

In contrast to that, a process for preparing TEDA as described in the US-A 2,937,176 (see enclosed) in particular column 3, 2nd paragraph) is generally performed at entirely different reaction parameters. The temperature is in the range of 300 to 500°C, preferably from 330 to 360°C. Usually, the reaction is carried out at atmospheric pressure and in the gas-phase (see column 3, 3rd paragraph). Most important, different educts are employed, since no ammonia is used for the production of TEDA.

Again, the applicant's independent claim 11 is drawn to a process for preparing triethylenediamine (**TEDA**), which is a **cyclic tert.-amine** by the reaction of ethylenediamine (EDA) and/or piperazine. This reaction is not a condensation reaction. Moreover, it is surprising that in contrary to Ashina a selectivity improvement of a **cyclic tert.-amine** is achieved. By the process of Ashina a selectivity improvement of a **non-cyclic sec.-amine** is achieved.

Consequently, Ashina gives no hint to the object of claim 11 and therefore, new claim 11 is not obvious in view of Ashina.

A statement that modifications of the prior art to meet the claimed invention would have been “obvious to one of ordinary skill in the art at the time the invention was made” because the references relied upon teach that all aspects of the claimed invention were individually known in the art is not sufficient to establish a *prima facie* case of obviousness without some objective reason to combine the teachings of the references. *Ex parte Levengood*, 28 USPQ2d 1300 (Bd. Pat. App. & Inter. 1993). See MPEP § 2143.01 IV. “[R]ejections on obviousness cannot be sustained by mere conclusory statements; instead, there must be some articulated reasoning with some rational underpinning to support the legal conclusion of obviousness.” *KSR International Co. v. Teleflex Inc.*, 82 USPQ2d 1385, 1396 (2007) quoting *In re Kahn*, 441 F.3d 977, 988 (Fed. Cir. 2006). Furthermore, the Examiner cannot selectively pick and choose from the disclosed parameters without proper motivation as to a particular selection. The mere fact that a reference may be modified to reflect features of the claimed invention does not make the modification, and hence the claimed invention, obvious unless the prior art suggested the desirability of such modification. *In re Mills*, 916 F.2d 680, 682, 16 USPQ2d 1430 (Fed. Cir. 1990); *In re Fritch*, 23 USPQ2d 1780 (Fed. Cir. 1992). Thus, it is impermissible to simply engage in a hindsight reconstruction of the claimed invention where the reference itself provides no teaching as to why the applicant’s combination would have been obvious. *In re Gorman*, 933 F.2d 982, 987, 18 USPQ2d 1885, 1888 (Fed. Cir. 1991).

Furthermore, contrary to what the Examiner has alleged, there would be reason for a person of ordinary skill in the art to use a molar ratio of SiO₂/Al₂O₃ less than 1400 when Frauenkron

specifically states that the minimum ratio is 1400. For the above reasons, this rejection should be withdrawn.

Assuming arguendo that the Examiner made a prima facie case of obviousness, the applicant believes that the declaration executed April 1, 2009 rebuts this rejection. In particular experiment 1 shows surprising effects of the subject matter of the present invention over Frauenkron and Ogawa.

In experiment 1, Dr. Ahrens had prepared two H-ZSM-5 catalysts, having a SiO₂/Al₂O₃ molar ratio of 1000 to 1. The catalysts were dried overnight at 120°C and subsequently calcined at temperatures of 500°C for 5 hours. Catalyst 1 (comparative example) was directly used in a process for preparing a triethylenediamine (TEDA) by reaction of ethylenediamine (EDA) and piperazine (PIP) according to claim 11.

Catalyst 2 (according to the invention) was treated with water vapor at 175°C for 24 hours. Subsequently catalyst 2 was used in a process according to claim 11.

The results were shown in table 1 of the declaration.

The treatment according to the invention leads to an increase of the EDA and PIP conversion from 97% (EDA)/49 % (PIP) to 99 % (EDA)/50% (PIP). Moreover the TEDA selectivity increases from 93 % up to 95 %. This surprising effect could not have been foreseen. The applicant has shown unexpected results and for the above reasons, this rejection should be withdrawn.

In view of the above amendment, applicant believes the pending application is in condition for allowance.

A three month extension has been paid. Applicant believes no additional fee is due with this response. However, if a fee is due, please charge our Deposit Account No. 03-2775, under Order No. 13156-00177 from which the undersigned is authorized to draw.

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Respectfully submitted,

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Enclosure: 1) U.S. Patent No. 2,937,176

2) ULLMANN' S ENCYCLOPEDIA OF INDUSTRIAL CHEMISTRY; PAGE 5